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ANNUAL REPORT

COMBUSTION MECHANISMS OF SOLID PROPELLANTS

By

E. W. Price, R. K. Sigman and R. R. Panyam

Prepared for

OFFICE OF NAVAL RESEARCH
ARLINGTON, VIRGINIA 22217

Under

Contract N00014-79-C-0674

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REPORT DOCUMENTATION PAGE

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1. REPORT NUMBER	2. GOVT ACCESSION NO. AD-A105 163	3. RECIPIENT'S CATALOG NUMBER
4. TITLE (and Subtitle) COMBUSTION MECHANISMS OF SOLID PROPELLANTS		5. TYPE OF REPORT & PERIOD COVERED ANNUAL TECHNICAL REPORT, 1 Aug 1979 - 31 July 1980
		6. PERFORMING ORG. REPORT NUMBER
7. AUTHOR(s) E. W. Price, R. K. Sigman ~~and~~ R. R. Panyam		8. CONTRACT OR GRANT NUMBER(s) ONR Contract No. N00014-79-C-0674
9. PERFORMING ORGANIZATION NAME AND ADDRESS GEORGIA INSTITUTE OF TECHNOLOGY SCHOOL OF AEROSPACE ENGINEERING ATLANTA, GEORGIA 30332		10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS 12 31
11. CONTROLLING OFFICE NAME AND ADDRESS OFFICE OF NAVAL RESEARCH ARLINGTON, VIRGINIA 22217		12. REPORT DATE July 1981
		13. NUMBER OF PAGES 28
14. MONITORING AGENCY NAME & ADDRESS (if different from Controlling Office) POWER PROGRAM OFFICE OF NAVAL RESEARCH ARLINGTON, VIRGINIA 22217		15. SECURITY CLASS. (of this report) UNCLASSIFIED
		15a. DECLASSIFICATION/DOWNGRADING SCHEDULE

16. DISTRIBUTION STATEMENT (of this Report)

Approved for public release; distribution unlimited.

17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)

Approved for public release; distribution unlimited

18. SUPPLEMENTARY NOTES

19. KEY WORDS (Continue on reverse side if necessary and identify by block number)

COMBUSTION, SOLID PROPELLANTS, ROCKETS, THEORY, SANDWICH BURNING

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

See next page.

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20. ABSTRACT

Investigation of mechanisms of combustion and combustion zone microstructure continued, using ammonium perchlorate-hydro-carbon binder sandwiches and the quench-burning method to obtain high resolution measurements. Binder-thickness deflagration limits were also determined, and work started on measurement of burning rate vs binder thickness. On the basis of results to date, a modified picture of the combustion zone is proposed that differs sharply from the classical model, particularly in those regions of the sandwich surface that are close enough to the AP-binder interface to correspond to processes in solid propellant combustion.

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OF SOLID PROPELLANTS

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by

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ONR Contract N00014-79-C-0764 July 27, 1981

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ACKNOWLEDGEMENTS

The research reported was sponsored by the Power Branch, Office of Naval Research. Technical monitor of the project is Dr. R. S. Miller. The authors appreciate the support of ONR in this research, and the enlightened management of the project typical of ONR.

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INTRODUCTION

Advances in prediction and control of combustion characteristics of heterogeneous propellants are severely limited by a lack of knowledge about structure and rate-controlling processes of the combustion zone. The marginal state of knowledge is due in large part to the microscopic dimensions and chaotic nature of the propellant and combustion zone structure. The chaotic microstructure has often been eliminated in past and present research by studies of propellant samples made from laminated sheets of oxidizer and binder, i.e., sandwiches. Real-time observations of edge-burning sandwiches can be made, with the location of the laminae, interfaces, and flame elements being established with fair accuracy. In addition, interrupted burning tests provide "post mortem" information about conditions in the condensed phase and burning surface to a much higher dimensional accuracy than observations during burning, because the quenched samples can be studied in optical and electron microscopes. From such studies, it has been possible to establish the combustion details on much less speculative grounds, and the emerging picture for propellants with ammonium perchlorate (AP) oxidizer and hydrocarbon (HC) binders is significantly different from the generally accepted models of combustion.

The present report summarizes studies of AP/HC binder sandwiches in the period 1 August 1979 - 31 July 1980, with some review of previous work to establish perspective. These current studies consisted of combustion photography and quench-burn tests on sandwiches with tapered binder laminae, i.e., tests in which the binder thickness was graduated from about 10 to 150 μm (wedge). This approach was started in a previous program,[1,2] and was continued here with emphasis on thin binder portions of the sandwiches (which are more relevant to propellants, and give combustion behavior different from behavior of the usual "thick-binder" sandwiches used in most other studies). The report describes quench profiles obtained over a range of binder thickness, pressure, and type of HC binder. Also reported are data on deflagration limits of sandwiches, and an heuristic argument relating observations to structure of the combustion zone.

EXPERIMENTAL METHOD

The experimental methods for preparing and testing sandwiches are described in a previous report (Ref. 1). Fig. 1 shows the sketches of tapered sandwiches. Fig. 1a shows a sample that has been burned down from a tapered edge, and Fig. 1b shows a sample that has burned down from the uniform thick edge. These particular examples were chosen because they illustrate the deflagration limit effect that occurs for thin binders. At pressures below the AP self deflagration limit, the sample in Fig. 1a burns down until quenched by rapid depressurization of the test vessel, but a portion of the "ignited" edge where the binder thickness is < 40 µm fails to burn. The sample in Fig. 1b burned down uniformly from the ignited thick-binder edge until the binder had decreased to 40 µm or less, and then went out spontaneously. The tests illustrated by Fig. 1a were used to obtain details of the surface profile vs binder thickness, while tests illustrated by Fig. 1b were used to determine deflagration limits. Details of the tests are summarized as follows:

All sandwiches were made in the same manner, by dry-pressing granular AP into 1.27 mm thick sheets and curing a binder lamina in place between two sheets. The AP used was Kerr-McGee 99.7% pure ground and screened to unimodal particle size in the 75 to 120 µm size range; pressing was at 165 MPa for 60 min. Binder ingredients were conventional propellant grade, provided by the Naval Weapons Center (PS, PBAN, HTPB, CTPB). Binder thickness was controlled by using a spacer between the AP sheets on one edge of the sample, and no spacer on the opposite edge. Actual thickness was determined from the quenched samples rather than from control of initial sample dimensions.

In tests, the samples were mounted in a nitrogen-pressurized chamber with vent at the top. The samples were ignited on the top edge with a hot wire and igniter paste. When controlled sample quenching was desired, it was accomplished by rapid depressurization ($\sim 3 \times 10^3$ MPa/sec) due to blow-out of a plastic burst diaphragm. Samples were subsequently coated with an evaporated film of gold, and examined in a scanning electron microscope (SEM). In the case of deflagration limit tests, several measurements were made of binder thickness along the quenched surface, using pictures from the SEM. In tests with combustion photography, a conventional nitrogen-flushed combustion chamber was used, and 16 mm motion pictures were taken at 2200

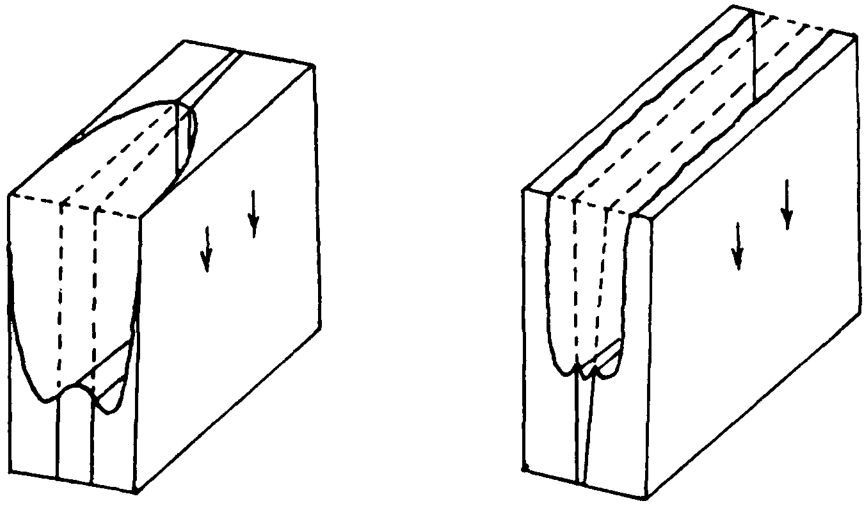
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Fig. 1 Quenched tapered binder sandwiches:
a) Sandwich burned on a tapered edge at 2 MPa and quenched by rapid depressurization.
b) Sandwich burned from the thick edge at 2 MPa and allowed to quench spontaneously.

frames per second with a picture width corresponding to 1 cm of the object plane.

Exploratory tests were also made to determine the possibility of obtaining burning rate in sandwich tests. This is difficult at most rocket motor pressures because the recessed nature of the leading edge of the burning front prevents accurate position determination from photographs during burning. Preliminary tests were run on "wired" sandwiches (Fig. 2) which would indicate passage of the burning front at intervals during burning. No successful tests were achieved, but work is continuing.

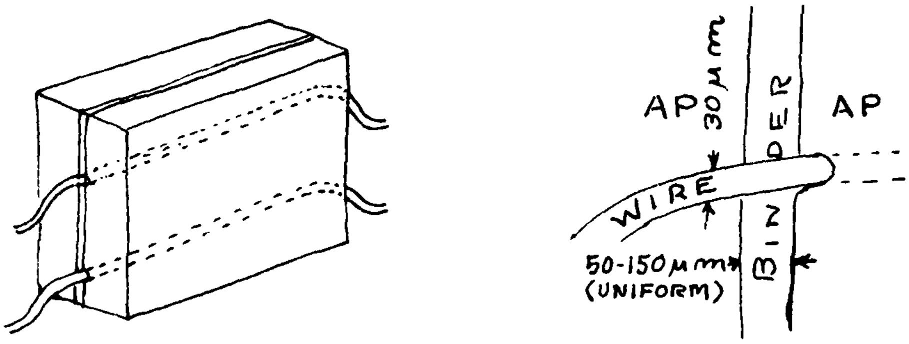

Fig. 2 Wired sandwiches for burning rate measurements.

RESULTS

As indicated above, the tests results consist primarily of quenched samples, which are studied by microscopic methods to determine surface profiles, surface details and binder thickness. The relation of these results to test conditions, and to general understanding of combustion mechanisms provide the basis for a qualitative theory of combustion presented in the Discussion.

Burning Surface Profiles

Observation of surface profiles was primarily a confirmation of results observed previously,[1,2] and reaffirmed that profiles are critically dependent on binder thickness. A sketch of profile trends is shown in Fig. 3. In anticipation of later discussion, it is relevant to note both classical and newly recognized features of profiles.

The most universally reported feature of surface profiles is the trend with pressure. Below the AP deflagration limit (parts a and d of Fig. 3), the

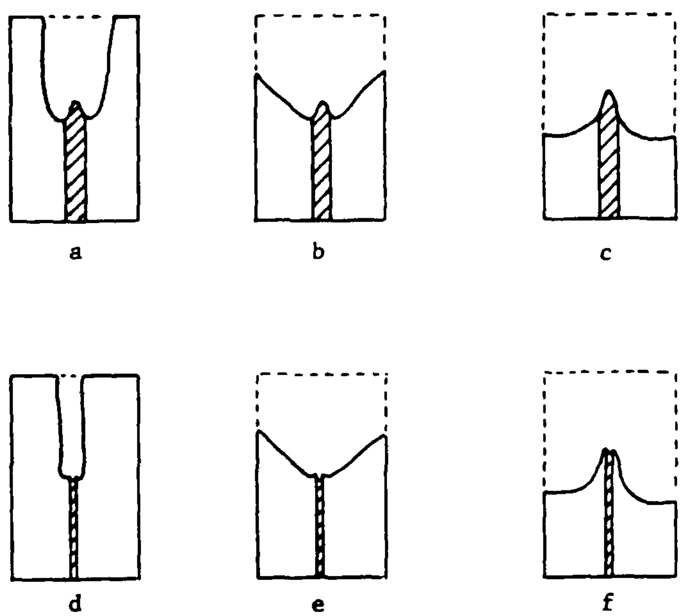

Fig. 3 Cross-section profiles of sandwich burning surfaces (sketched from quenched samples):
a) Low pressure (~ 2 MPa); b) Intermediate pressure (~ 5 MPa); c) High pressure (~ 8 MPa); d, e, f Same as a, b and c, but thin binder (~ 50 μm).

burning proceeds down the binder lamina, consuming an amount of AP roughly commensurate with a stoichiometric ratio to the binder. At intermediate pressure (parts b and c of Fig. 3), the AP deflagrates at its own characteristic rate, with the profile piloted by a faster-burning interfacial region at the "sandwich" burning rate (when the characteristic AP burning rate is known, the slopes of the "V" profile can be used to calculate the sandwich rate). At high pressure the AP deflagration rate exceeds the sandwich rate, with the result that the binder lamina protrudes above a flat AP surface (parts e and f of Fig. 3).

A more recently established feature of sandwich profiles is the trend with binder thickness. A result noted in limited tests in Ref. 3 - 5, and many tests in Ref. 1, 2 is the change from protruding to recessed binder surfaces as binder thickness is reduced (Fig. 3 a - c vs Fig. 3. d - f). This trend has been consistently supported in current tests as well, although there is some quantitative difference with different binders. These trends are currently being studied with families of sandwiches with uniform thickness, to determine consistency of results at each binder thickness.

A relatively recent[1,6] (and ongoing) observation about surface profiles concerns the details of the AP profile near the AP-binder interfaces. Early papers reported the leading edge to be at the interface, but the work reported in Ref. 3 showed that the leading edge of the AP was 20 - 100 μm out from the interfaces as implied in Fig. 3. Subsequent studies using sufficiently high resolution methods consistently confirmed this result and extended it to a wide range of test conditions (Ref. 1,2,4-6). In some instances it was obvious that the effect resulted from retardation of the AP regression rate near the interfaces by a flow of molten binder onto the AP.[3,7,8] It is only in recent work[1,2,6] that it became clear that the retardation of the AP regression rate is present even in the absence of binder melt flow, and with all binder thicknesses at all pressures. This has been established in the present studies for PBAN, PS, and HTPB binders, for binder thicknesses from 20 - 150 μm, and pressures from 13.8 MPa down to the sandwich low pressure deflagration limit. This point will be illustrated in the next subsection in connection with results concerning other attributes of this region of the burning surface.

Smooth Bands

In the previous report[1,2] it was noted that the surface quality of the quenched AP laminae in sandwiches (and propellants) was different near the AP-binder interface than elsewhere on the samples. While the majority of the AP surface exhibited the surface undulations and froth residue typical of AP self deflagration, the region near the interface plane exhibited an almost "glossy" appearance in scanning electron microscope pictures (Fig. 4). Current work has confirmed that this "smooth band" adjoining the interface plane is coincident with the region of retarded regression of the AP surface (Fig. 5), indicating that these are two separate manifestations of a single attribute of the local AP deflagration that is different from that elsewhere on the sample, a difference linked to the proximity of the binder lamina.

The possibility that the smooth band effect is a result of binder melt flow seems to be eliminated because in results to date the band is persistently present, even in situations not conducive to flow. These situations include: relatively dry-burning binders such as polysulfide; thin binders that burn in a recessed manner (Fig. 5,6); samples burned upside down so the gravitational effect resists flow onto the AP surface. Smooth bands and locally retarded AP regression occur in all these situations.

Another possibility that was considered as a possible cause of smooth bands and retarded regression was that, during sandwich fabrication, some binder or fuel-like ingredient might diffuse into the AP lamina to give a modified layer with modified deflagration characteristics. This possibility was rejected on several grounds. There was no direct evidence of fuel diffusion. If it did occur, it was reasoned that it would <u>increase</u> regression rate, not retard it. Further, the same effects (smooth band, retarded rates) seem to have occurred consistently also in earlier testing[3-5] where the AP laminae were cut from large single crystals that would be nonporous. In addition, tests were run in the present study using AP laminae pressed to lower density, and laminae pressed from very fine (5 μ m) AP. These modifications had no noticeable effect on the smooth band phenomenon. Thus it is concluded that fuel permeation is not responsible for the smoothness and rate effects.

An earlier critical test regarding smooth bands involved interruption of burning of a sandwich in which the fuel lamina was replaced by a sheet of

a b

Fig. 4 Scanning electron micrographs of quenched tapered sandwiches showing characteristic surface profiles and surface details such as smooth bands. Binder thickness in these pictures ranges from 70 μ m (left) to 10 μ m (right).

4a AP/CTPB/AP sandwich quenched from 1.38 MPa. Top shows most of sample, lower shows smooth band, with recessed binder at top.

4b AP/PS/AP sandwich quenched from 4.1 MPa. Top shows the thin binder end of the burning surface, lower shows smooth bands on each side of binder, typical AP self-deflagration surface further from binder. (PS binder gives little or no evidence of melt or flow.)

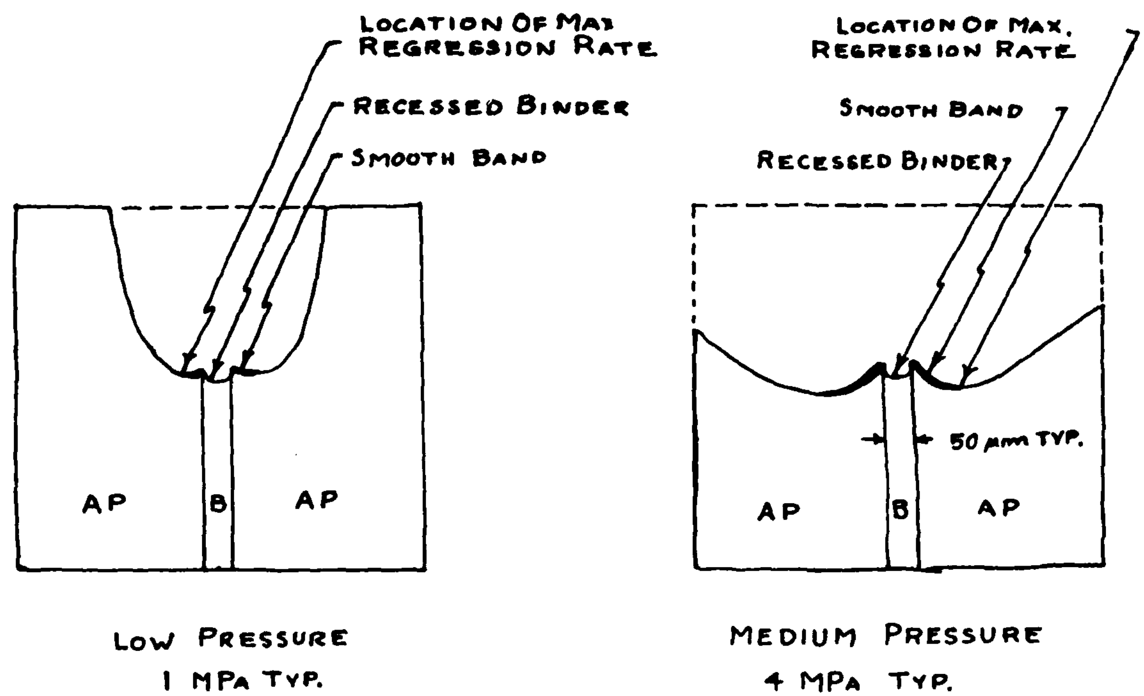

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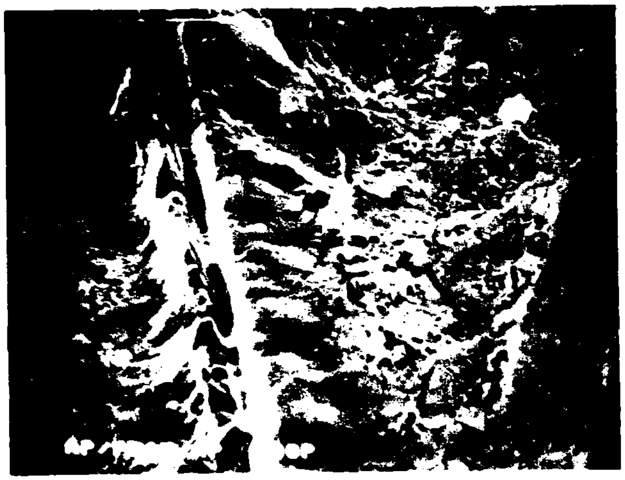
Fig. 5 Sketches of profiles of thin-binder sandwich sections, showing locations of characteristic features of profiles and burning surfaces. 5a Typical of 1 MPa; 5b Typical of 4 MPa.



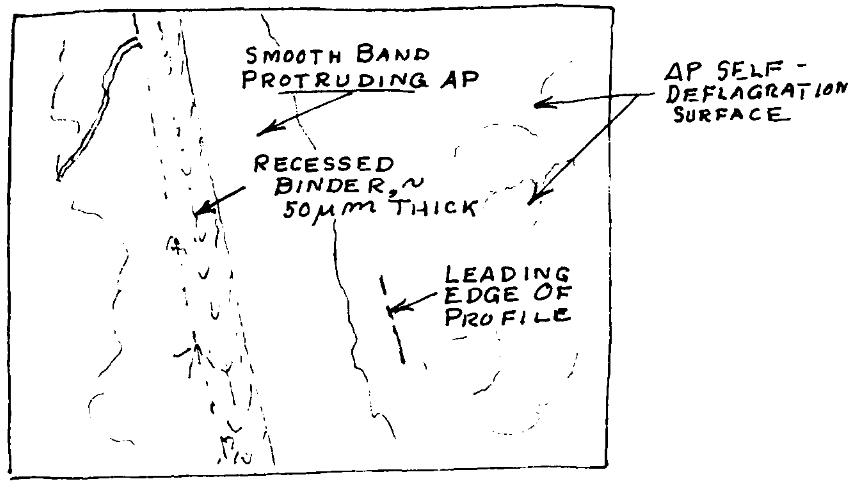


Fig. 6 Uniform-binder sandwich quenched from 4.14 MPa (AP/PBAN/AP, binder thickness 30 μ m). Note retardation of AP regression near the binder interface, and the relatively smooth quality of the surface in this region. Note region of maximum AP regression is near "outer edge" of smooth band.

of mica.[1,2] This test resulted in absence of the smooth band and of the retarded rate effect. This test was repeated in the present study with the same result. Mica was chosen originally because it is a non-reactive thermal insulator. Its use removes the AP-binder flame and minimizes heat transfer in the non-AP lamina. Tests are in progress on sandwiches with non-reactive, thermally conductive laminae (gold leaf), to try to determine to what extent the "mica results" are due to removal of the AP-binder flame and to what extent they are due to changes in heat conduction. Preliminary results indicate that the smooth band and retarded AP regression are present when a 14 μm thick gold lamina is used.

Binder Thickness/Low Pressure Deflagration Limits

Tests such as that illustrated by Fig. 1b yielded a set of measurements of binder thickness from each spontaneously quenched sample, an average value, and a range of values. These results are shown in Fig. 7 as a graph of binder thickness vs test pressure, for three different binders. The results indicate the following features:

1. There is a minimum binder thickness for self deflagration of sandwiches, which is relatively insensitive to pressure down to about 0.4 MPa. This minimum value of thickness is about 20 μm for PBAN binder, and 30 μm for PS and HTPB binder.

2. Below about 0.4 MPa, the limiting thickness of binder increases rapidly with decreasing pressure.

3. Self quenches occurred in some tests at pressures above the stated AP self deflagration limit.

4. The range of binder thickness in any one test sample was fairly large, especially with PS binder. This data scatter, which is common to most flammability measurements in combustion science, seems nonetheless to establish items 1 - 3 above beyond doubt for this experiment.

There also appears to be a significant but small systematic trend in binder lamina thickness vs pressure in the $>$ 0.4 MPa range which may also prove useful in mechanistic interpretations, but reproducibility probably does not warrant speculation now.

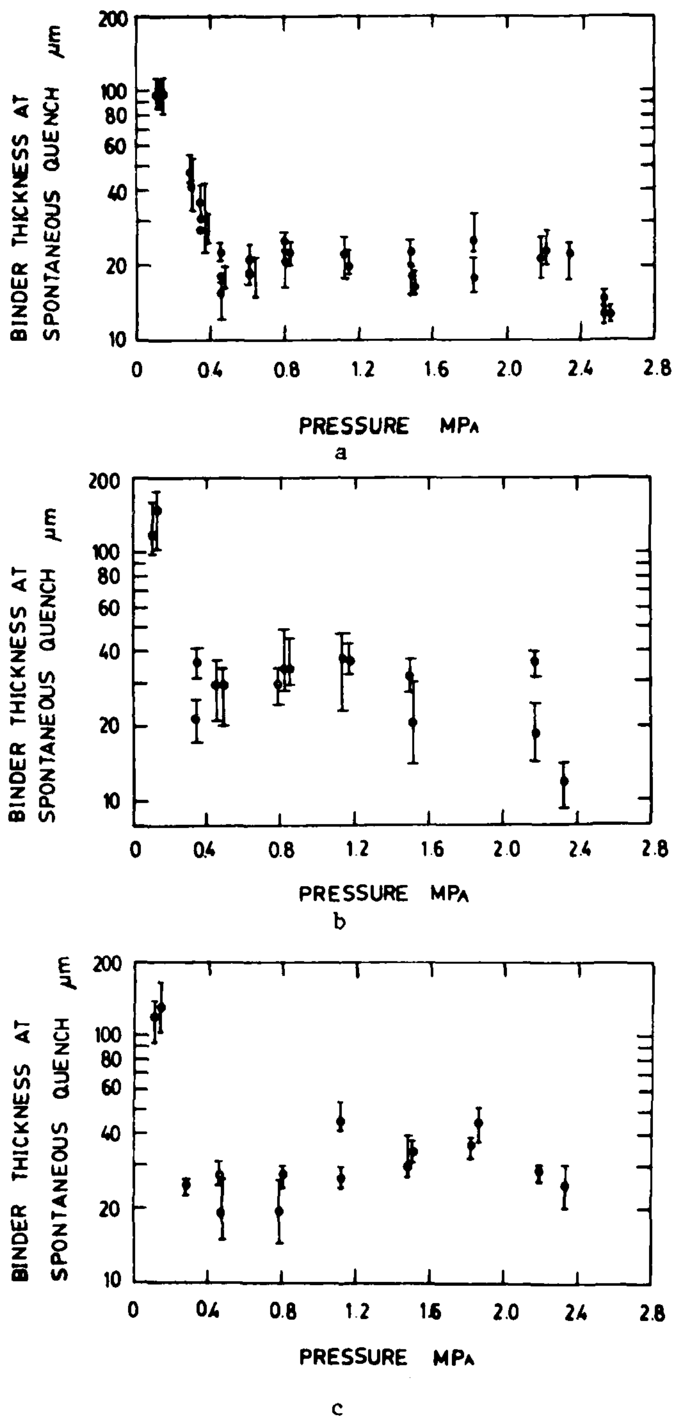


Fig. 7 Deflagration limits for tapered binder sandwiches burned at constant pressure and spontaneously quenching at the indicated binder thickness: a) PBAN binder; b) PS binder; c) HTPB binder.

THEORETICAL CONSIDERATIONS

Ideally, one would like to use the experimental results for guidance in formulation of an analytical model of sandwich burning, and then test the detailed agreement between model and experiment. In this, one would be guided by previous efforts and hope for refinement of those results. However, it is doubtful that an analytical model of AP-HC sandwich burning can be constructed now that is both realistic and tractable. That would seem to be a longer range goal, that is one of the mileposts on the trip to propellant modeling. On the other hand, there is a great deal known about the combustion mechanisms, and it appears logical to construct a qualitative description that can serve as a basis for interpretation of current experimental results, for design of future experiments, and for constructing analytical models.

The more generally accepted views of sandwich burning mechanisms are embodied in Fig. 8. The oxidizer (AP) burns with its own flame, standing close to the surface. In addition, there is substantial support for the view that an exothermally reacting melt layer is present at pressures > 2 MPa, i.e., above

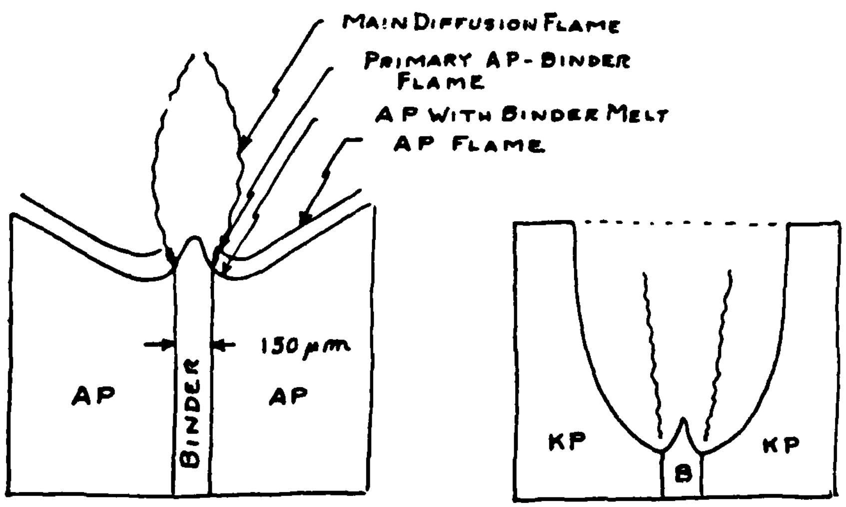


Fig. 8 Conventional view of the combustion zones of AP/HC, and KP/HC sandwiches.

the AP low pressure deflagration limit. Relatively little is known about the AP flame below 2 MPa, although it is plausible to assume that it is present at lower pressures in regions where the AP-binder flame supplies sufficient heat. When the flame is absent, the AP probably decomposes endothermally by dissociative sublimation to NH_3 and $HClO_4$.

In the micro region close to the AP-binder interface, the oxidizer vapors and binder vapors mix quickly, to give the kinetically controlled "phalanx" flame proposed by Fenn.[9] There is no direct experimental evidence of the location or influence of the phalanx flame. In some respects it is analogous to the "primary" flame used by Beckstead[10] and others in modeling combustion of propellants. However the "primary" flame is defined arbitrarily as the portion of the oxidizer-fuel (O-F) flame closer to the surface than the AP flame. It is not clear whether this is more, or less, than the kinetically limited phalanx flame of Fenn (who was not considering a self deflagrating oxidizer). In any case, a portion of the O-F flame more distant from the surface is diffusion-limited, because of the longer diffusion paths for reactants at those locations. As noted by Summerfield,[11] the portion of the flame that is kinetically limited increases as pressure is reduced, because kinetic rates decrease more rapidly with pressure than diffusion rates do. In the case of AP sandwiches, the pressure dependence of the overall flame complex is further enhanced at low pressure by the tenuous nature of the AP flame.

The sketch in Fig. 8 shows a protruding binder lamina, and an AP profile that slopes up to a protruding binder. As noted earlier, the protrusion of the AP has been recognized for many years. However the reason for the protrusion has not been established, and only recently has it been recognized that the condition exists even when the binder is recessed. This question will be addressed later in this report, but to do so it is advantageous to go back and address first some more general questions about how sandwiches burn. This will be done by starting with the simpler situation, the burning of sandwiches with "non-selfdeflagrating" oxidizers. This is the situation assumed in most analytical models.[9,12-14]

Single Interface Burning

Consider first the burning of a "composite" consisting of a slab of binder

against a slab of non-self deflagrating oxidizer, ignited at an exposed edge of the interface plane. The flame is pictured as in Fig. 9, consisting of a leading edge that is a kinetically limited (pre-mixed) flame, with a transition to a trailing diffusion-controlled flame further from the interface. Both analytical and experimental evidence of the structure of this flame complex is speculative, involving as it does microscopic dimension and uncertain information about transport and kinetic properties in the region. For convenience, a region of the leading edge of the flame complex is identified as a "propagation velocity controlling" (PVC) region, i.e., that part of the flame that contributes to pyrolysis of the solids at the leading edge of the surface profile. Because it is difficult to model the PVC region accurately, it is also difficult to explain the surface profile accurately—but accurate experimentally determined profiles may provide good tests of the validity of models. For the present purposes, the classical assumption will be adopted, that the PVC region includes both the premixed flame and part of the diffusion flame, with the relative contributions from the two parts being dependent on pressure, etc.. The contribution of an AP decomposition flame, if any, will be considered later. Thus single interface burning is described as a PVC flame that determines the leading edge velocity and profile, with a trailing flame that sustains burning of the ingredients further out from the interface.

Sandwich Burning

Most experimental work on burning of laminated systems has been done on sandwiches, consisting of a fuel lamina between two laminae of oxidizer. If the fuel lamina is thick enough, such a system will burn with two substantially independent interface flame fronts as in Fig. 10. Such flame systems would presumably interact with each other only at the trailing edge of the lamina. Most investigators have chosen fuel laminae that are thin, in the sense that the overall sample stoichiometry is fuel lean. Under these conditions the burning proceeds down between residual walls of oxidizer, with the fuel lamina protruding moderately from the bottom of the slot. The extent of fuel lamina protrusion, and hence of separation of the PVC regions, varies with the fuel lamina thickness (Fig. 11), as well as fuel pyrolysis characteristics, pressure, etc.

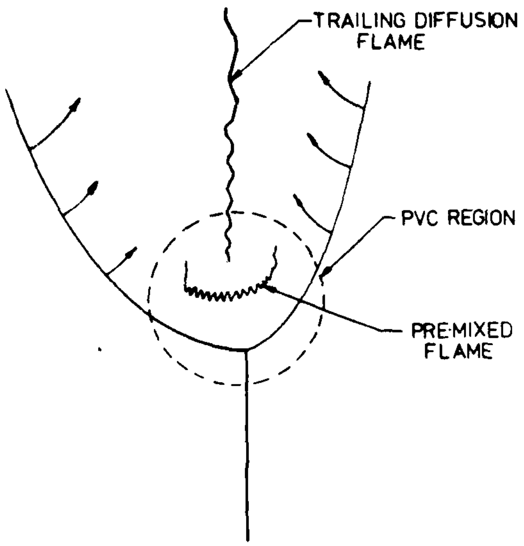
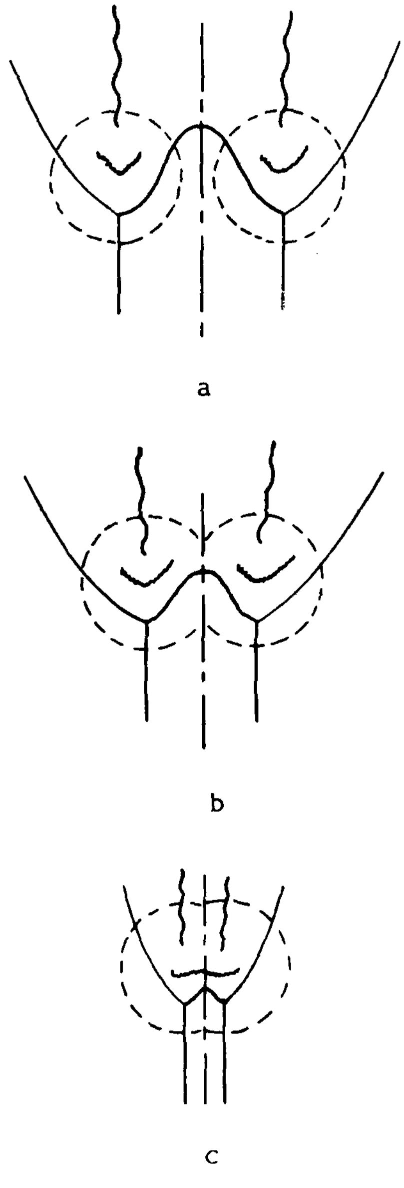


Fig. 9 Model of burning down the interface between an oxidizer and a fuel slab.



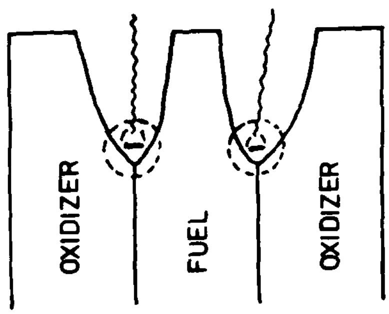


Fig. 10 Combustion of a sandwich with thick fuel layer (two non-interacting PVC flames).

Fig. 11 Effect of reducing fuel lamina thickness (showing onset of PVC region interaction): a) intermediate thickness; b) thin binder; c) very thin binder.

In order that the results of studies on sandwiches will be relevant to combustion of heterogeneous solid propellants, one must examine what aspects of sandwich burning can in fact approximate some aspect of propellant combustion. Evidently, the first requirement is that behavior be simulated and examined on the same microscopic dimensional scale as the propellant combustion zone microstructure. It apparently does not matter whether the sandwich is fuel lean or fuel rich, so long as that does not affect the PVC region.* But if the binder lamina is too thick, the interaction of PVC region pairs may be prevented, or excessive binder melt may flow into the PVC region. Since the actual dimensions of the PVC regions are not known, their dimensions will be as much a goal of experiments as a guide to design of them. At the outset, the characteristic dimensions of the propellant microstructure suggest that the spatial domains of relevance in the solid are from 0 to about 200 μ m in the oxidizer, with typical thickness of binder structural elements being from 0 to 50 μ m. Consequently the present studies have addressed primarily sandwich burning results with binder thickness in the 0 - 70 μ m range with larger thickness being used primarily to provide continuity with that large portion of previous sandwich burning research that used binder thicknesses > 70 μm.

The foregoing strategy for relevance does not settle the question of dimensions of the PVC region in a fundamental sense; it only sets limits on dimensions of the part of the PVC region that actually have an opportunity to occur in the propellant combustion zone. This is closely related to the issue of proper modeling of propellant combustion and the effect of particle size on propellant combustion. The issue will be examined here in the restricted scope of the sandwich combustion zone.

<u>The Sandwich Flame Dimensions</u>

When the binder lamina is fairly thick, diffusion processes lead to heat and oxidizer diffusion** laterally out of the PVC regions into a region of the binder lamina and to fuel flow that ultimately is convected away <u>between</u> the

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* A somewhat larger region may be involved in other aspects of burning such as oscillatory combustion.

** Heat diffusion via both solid and gas phase.

PVC regions (Fig. 11a). As the binder lamina is taken thinner, its tip retracts (Fig. 11b, c), while the PVC regions come closer together, and the loss of heat and oxidizer from between the PVC regions decreases until the PVC regions merge into a single combustion wave (Fig. 11c). One might anticipate that this more conservative flame would burn more rapidly into the sandwich, and that the binder thickness where the increase in burning rate showed up would be an indicator of dimensions of the PVC region. While there is only minimal data available on burning rate of sandwiches, Fig. 12 is a rough sketch of trends reported in Ref. 12-17, showing burning rate as a function of binder thickness for potassium perchlorate and ammonium perchlorate systems, adjusted to a pressure of 2.0 MPa. As the binder thickness is reduced from a large value, a burning rate increase begins at about 1 - 2 mm for KP. A similar but less conspicuous effect is observed for AP sandwiches, with the binder thickness being 0.5 - 1.0 mm. Thus the two PVC regions begin to affect each other when interfaces are 0.5 - 2 mm apart, implying a fuel-side width of the PVC region of a single interface flame of about 250 - 1000 μ m (pressure dependent).

The broken-line parts of the burning rate curves in Fig. 12 were inserted without any direct experimental data, to indicate an expected trend to decreasing burning rate with decreasing binder thickness. This trend is due to increasing lateral losses on the oxidizer sides of the flames as the flames burn down an increasingly narrow slot in the sample. In this thickness range the two flames are presumably fully merged on the fuel sides, so that no compensating "gains" occur on the fuel side to keep the burning rate up as fuel thickness is reduced, so the exterior loss trend dominates. The validity of this postulated trend of burning rate with binder thickness is supported not only by the above reasoning, but also by extrapolation of experimental results of Ref. 14 and by the present observation of minimum thickness for sustained burning of sandwiches (Fig. 7). The thickness scale of the curves in Fig. 12 was chosen on the basis of reconciliation of experimental sources (Ref. 12-17), with the dominance of outer "losses" over inner "gains" occurring when binder thickness is reduced below 100 μ m (AP) - 300 μm (KP).

The trend of the thin-binder ends of the curves in Fig. 12 terminates at the deflagration limit. No limits were found in the literature for KP sandwiches, but those for AP are reported here in Fig. 7. Tests on KP sandwiches with 125 μ m PBAN binder were attempted, but ignition could not

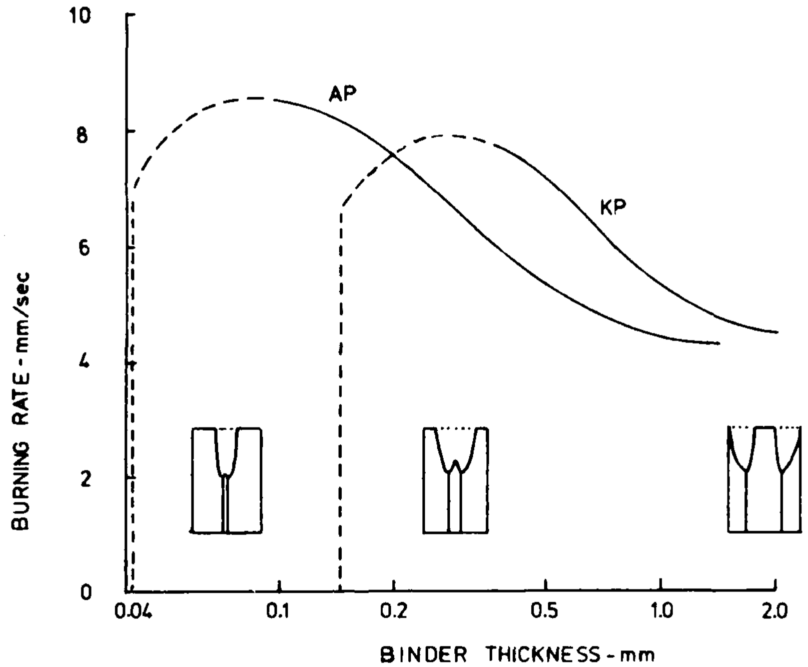

Fig. 12 Trend of sandwich burning rate with binder thickness.

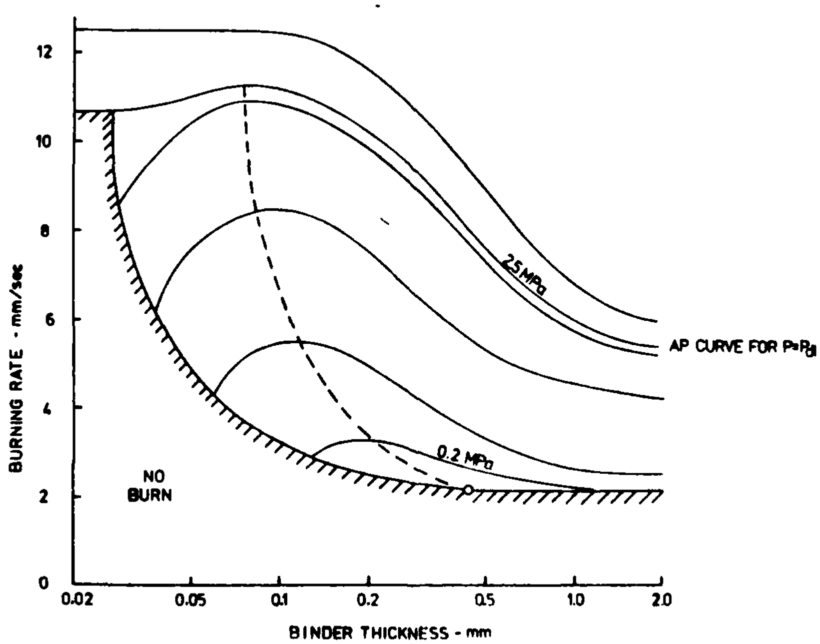

Fig. 13 Trend of sandwich burning with binder thickness and pressure, for oxidizer that self deflagrates above 2.5 MPa.

be achieved at any pressures tested (2.2 MPa to 6.9 MPa). It should be noted that the curves in Fig. 12 are only estimated. The results in the literature represent a variety of experiments, often only superficially described, and with results that either should not, or could not, be quantitatively reconciled. However, they do seem to provide a reasonable basis for the qualitative trends described here. In combination with the interpretation of the results, a basis is provided for estimates of the dimensions of the PVC regions and the burning surface profiles (sketched in Fig. 12 and discussed shortly).

The foregoing can (albeit recklessly!) be extended to a general picture of sandwich burning as in Fig. 13. In this figure, the more gross contributions of AP self-deflagration (neglected to this point) are introduced as a change in burning rate trends with pressure, and a change of the AP sandwich deflagration limit to zero thickness above 2.5 MPa. Obviously, the argument needs to be tested by some systematic burning rate measurements, but present results suggest a PVC region that is between 25 and 100 μ m wide on the fuel side, with a similar stand-off distance from the surface.

Burning Surface Profiles

To date, no realistic analytical models of sandwich burning have been developed, and even the simpler process of single interface burning suffers from lack of complete description of the PVC region, even for KP oxidizer. Thus most of the generalizations reached in the present work regarding surface profiles can be discussed in only qualitative terms. The big uncertainty seems to be the self-deflagration behavior of the oxidizer. When the AP deflagrates, it produces a flame close to the burning surface, that is believed to contribute appreciably to heat conduction to the burning surface. But under what conditions does that flame occur? Does it occur in the PVC region? Does the diffusion of fuel species enhance or suppress the AP flame? How does the lateral heat flow in the laminae affect the AP flame? None of these questions can be answered decisively at present, yet answers would be necessary for formulation of a combustion model.

There was one striking contrast between the present results with AP sandwiches and those predicted and observed with single interface models and KP-fuel slab tests, i.e., the retardation of the oxidizer regression adjacent to

the interfaces. At high pressure (e.g., 10 - 14 MPa) this can be regarded as a case of dominance of the AP self-deflagrating rate over the interface burning rate. This interpretation is not likely to explain the extreme protrusion of both binder and AP that actually occurs (Fig. 3c, 3f, 6). It is postulated that the extreme protrusion is due to failure of the AP flame to maintain itself near the interface. This hypothesis is supported not only by the profiles, but also by independent evidence that the AP self-deflagration flame is only marginally stable in the absence of fuel lamina,[18-19] with the low pressure deflagration limit being very sensitive to impurities, heat losses, and ambient sample temperature. In sandwich burning, the AP near the interface loses heat to the endothermic binder, an effect that would quench the AP flame, with corresponding reduction of heat transfer rate to the surface, reduction of regression rate, and a corresponding protrusion of the interface AP.

At low pressure, one may call on substantially the same argument. One may argue that the AP flame persists in favored locations even below the low pressure deflagration limit provided it is supported[20] by the AP-fuel flame. However, the AP flame would, as argued for the high pressure situation, fail to sustain in close proximity to the interfaces where losses to the endothermic binder lamina are excessive. This would explain the continued retardation of interface AP, even at low pressure. It may also explain the most striking observation of the present tests, the correspondence of the locations of the smooth band and the region of retarded AP regression adjoining the interfaces. We suggest that the smooth band is associated with the absence of an AP flame.

These rather speculative interpretations of persistent effects in the sandwich tests have fairly profound implications for propellant combustion, where the entire burning surface is made up of structural entities of the same dimensional order as smooth bands, retarded regression profiles and postulated AP flame quenched regions. In the meantime, the collected results give a clue as to flame zone structure and dimensions.

Deflagration Limits

It has been noted that the thin-binder sandwiches are very fuel lean, and below the AP deflagration limit the sandwiches burn down a narrow slot in the

sample. In burning down tapered sandwiches, the PVC flame is subject to increasingly high (proportional) heat loss as the binder thickness decreases and the slot becomes more narrow. The resulting reduction in PVC flame temperature would be expected to lead to reduced reaction rates, and ultimately to quenching. In the deflagration limit experiments, this situation is reached in each test. At pressures about 0.4 MPa, the quench occurs at a binder thickness that is relatively insensitive to pressure (Fig. 7). This result is interpreted here as implying that diffusion, rather than kinetics, is dominant in controlling either reaction rate in general, or the shape of the flame. The relative pressure independence of diffusion controlled burning then controls the approach to the deflagration limit. Kinetic considerations presumably become more dominant near the deflagration limit, due to the decrease in flame temperature in the narrowing slot. If this interpretation is correct, there is no way to determine from these results whether the moderate dependence of the deflagration limit on binder type is due to differences in kinetics, flame temperature, or fuel molecule transport properties. A more general conclusion is instinctively drawn from the trend in limiting binder thickness with pressure, i.e.,that the PVC flame is diffusion dominated down to 0.4 MPa At lower pressure the binder thickness deflagration limit becomes extremely pressure dependent. This behavior suggests that the PVC flame has become kinetically limited. However, these speculations may be premature, in view of the intricate flame complex that is suggested by the present studies.

DISCUSSION AND SUMMARY

The objective of the present research is to determine the rate-controlling steps in combustion of heterogeneous propellants, starting with AP-HC propellants. The term "rate controlling" is used in a broad sense here, to mean steps that control various aspects of propellant combustion, such as burning rate, dynamic response to pressure and flow oscillations, and single transient events such as ignition, DDT, and quenching (the controlling steps are not the same in all of these aspects of combustion). The immediate goal is less ambitious, i.e., to determine the structure of the steady state combustion zone, and how it depends on pressure, ingredient particle size, etc. The sandwich burning approach was adopted in order to obtain less ambiguous test results and easier interpretation of those results.

Because of the microscopic, two-dimensional nature of the combustion zone, real-time observations by combustion photography and other methods provide only indirect information, and resort has been taken to "post mortem" observations, i.e., high resolution microscopic examination of samples from quenched burning experiments (augmented by burning rate and deflagration limit experiments). Determination of the combustion zone microstructure involves reconciliation of the results of a variety of such experiments, and known principles of combustion science to produce a combustion theory that will stand the test of subsequent experiments, or correctly predict their outcome. The results to date are controversial enough to raise discussion and pose new issues and experiments. They are well founded enough to encourage continued investigation with the expectation that a new and more realistic theoretical model will result. They are preliminary, and hence subject to some extension and "quantification". The picture of the combustion zone that has emerged is shown in Fig. 14. The principal features are:

1. An exothermally reacting froth on most of the AP surface. The froth appears to be present everywhere that the multiple flame complex maintains a surface temperature as high as that present during normal AP self-deflagration.
2. An AP flame, close to the AP surface, present whenever the froth layer is present.

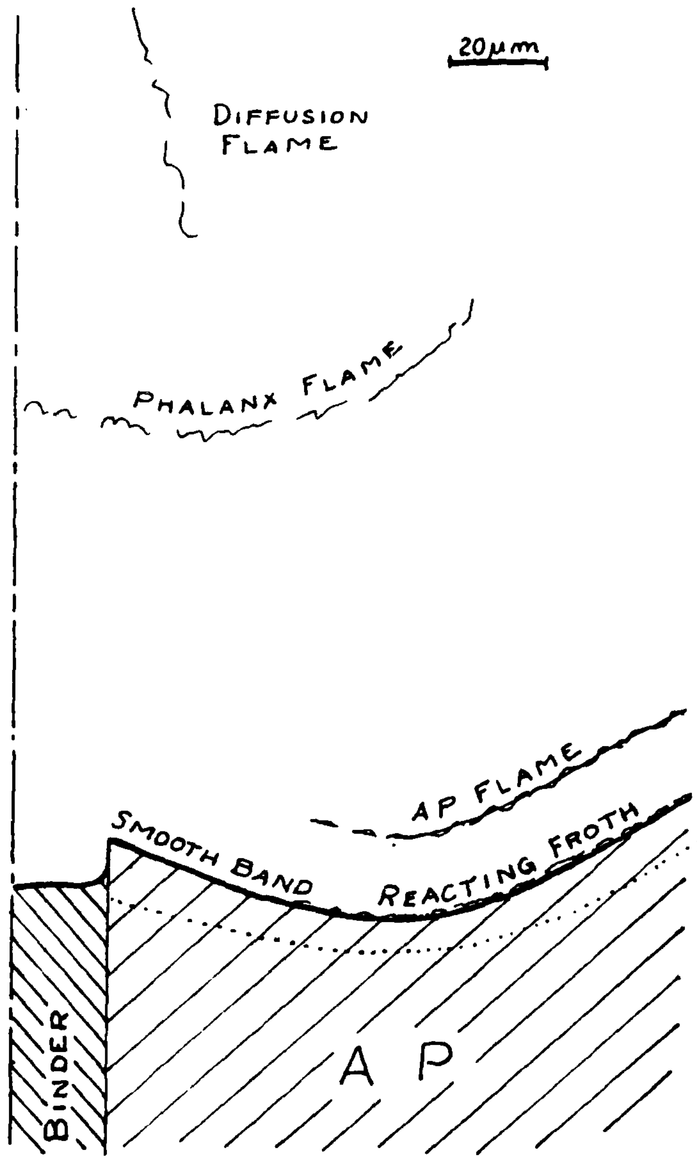

Fig. 14 Postulated combustion zone microstructure for thin-binder sandwiches.

3. A "broad", kinetically limited "phalanx" flame, more remote from the surface than the AP flame, but still affecting surface regression for some distance out from the AP-binder interface plane.

4. A diffusion limited flame still more remote from the leading edge of the burning front (one for each AP-binder interface plane?), absent when the binder is so thin that the fuel is all consumed in the phalanx flame.

5. A "smooth band" region on the AP surface adjoining the edge of the AP-binder interface plane; the reacting froth, and probably the AP flame are absent here, and AP vapors (i.e., NH_3, $HClO_4$, and other intermediate products) mix with fuel vapors and feed the phalanx flame.

6. A resulting surface profile with recessed binder (when not too thick), a feature that reflects the lower temperature required for decomposition of binder at the rates present. The AP regression is retarded in the smooth band region because of heat loss to the binder and resultant local quenching of AP exothermic reactions. The leading edge of the AP burning front is outside the smooth band, where the AP flame is less impeded by heat drain and supported by heat transfer from the phalanx flame.

7. Combustion zone dimensions are roughly as shown in Figure 14 typical of a pressure of 5 MPa.

At low pressure the phalanx flame is presumably further from the burning surface, and the AP flame may fail to be sustained by the phalanx flame. With sufficiently thin binder, the phalanx flame may consume all of the fuel (no diffusion flame). At high pressure (> 7 MPa), the AP self-deflagration rate is higher than the rate of the overall flame complex, so the region near the interface protrudes. The details of the flame complex, consequently, become less important to burning rate, but are probably still important to other aspects of combustion (e.g., in propellant combustion). The faster kinetics at higher pressure presumably cause the phalanx flame to move in closer to the surface, in the process limiting the amount of pre-mixed reactants available and increasing the contribution of the diffusion limited part of the flame. At high pressure, the net effect is a retardation of burning rate relative to the AP rate, so that the AP portion of the sandwich dominates burning rate.

In summary, the principal results of recent work are:

1. Further detailed verification of previously reported quench profiles vs pressure, binder thickness, and kind of binder. Particular emphasis was placed on verifying the details of smooth bands and retarded regression rates near the AP-binder interfaces. Efforts are now directed at obtaining quantitative data on these details of combustion zone microstructure.

2. Exploration of the effect of changes in "fuel" lamina material to clarify the mechanistic basis of smooth bands and retardation of regression rate (mica, gold, and AP-AP without any intervening lamina). This investigation is continuing.

3. Preliminary efforts at determination of burning rates of thin-binder sandwiches in the < 5 MPa domain where current methods and data are inadequate. The purpose of these experiments is to establish the curves in Fig. 13 on a factual basis. Efforts are continuing.

4. Systematic determination of the sandwich deflagration limit (binder thickness vs pressure) for three binders.

5. Development of a qualitative model for sandwich burning, as a preliminary to quantitative modeling.

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